Comparison of Nd^{3+} in $GdLiF_4$ and $YLiF_4$ by Fourier Spectroscopy

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Abstract

An investigation on the spectroscopic properties of Nd^{3+} doped $GdLiF_4$ (GLF), a new laser crystal, is reported. Our high resolution absorption and emission spectra for GLF are nearly identical to those of Nd:$LiYF_4$ (YLF) strongly suggesting that the two crystals are isostructural. The linewidths of Nd:GLF are independent of Nd concentration up to 4%.

Laser action of a new crystal Nd:$GdLiF_4$ is being reported for the first time by Zhang et al. at this conference[1]. The laser performance Nd:GLF is very similar to that of Nd:YLF. One advantage of the new material over YLF is that much higher Nd concentrations can be realized. Low resolution, room temperature absorption spectra[1] reveal essentially no difference in peak positions with Nd:YLF. Here we present a high resolution low temperature study in order to accurately determine the energy levels of Nd:GLF and to compare these with Nd:YLF levels determined by us using the same technique.

The high optical quality single crystals of Nd:GLF were grown by the modified Czochralski pulling technique. Two samples were nominally doped with 5 at.% and 1.3 at. % Nd. The actual Nd^{3+} concentrations are estimated to be about 4 at.% and 1 at.%, respectively. The Nd concentration of our YLF sample was nominally 3% but is estimated to be 1.1 at.%.

We have measured polarized, temperature dependent, transmission, and emission spectra from 500 to 22,000 cm^{-1} using a Bomem DA8 Fourier spectrometer. Resolutions up to 0.5 cm^{-1} were used in order to resolve all lines. One advantage of the Fourier technique for characterization of laser materials is its high frequency accuracy, being 0.004 cm^{-1} at 2000 cm^{-1} for the Bomem. Photoluminescence was excited by a multiline Argon laser.

Figs. 1 and 2 show the $^4F_{3/2} \rightarrow {}^4I_{11/2}$ photoluminescence spectra for both polarizations at a sample temperature of 80K. All emission spectra are normalized (maximum emission =1). This group contains the strong transitions used for the 1μm laser operation. The spectra appear nearly identical. The locations and relative strengths of the lines are very similar. The spectral widths in GLF are somewhat broader than those in YLF. The full widths at half maximum (FWHM) for both 1.047 and 1.053 μm lines are given in Table 1 for Nd:GLF and Nd:YLF. The widths in GLF are independent of Nd concentration within experimental error.

Table 1. Full widths at half maximum (cm^{-1}) of the main emission peaks in Nd:GLF and Nd:YLF at 80 and 300 K

crystals at 80 K	1.047μm π-polarization	1.053μm σ-polarization
1.0 at.% Nd:GLF	4.6	3.0
4.0 at.% Nd:GLF	5.0	3.5
1.1 at.% Nd:YLF	2.9	2.2
crystals at 300 K		
1.0 at.% Nd:GLF	18.0	15.4
4.0 at.% Nd:GLF	17.3	15.3
1.1 at.% Nd:YLF	12.4	11.1

Fig. 3 presents the $^4I_{9/2} \leftrightarrow {}^4F_{3/2}$ transmission and emission lines in π-polarization. These are transitions between the ground multiplet and the upper laser levels. The strongest line and the weak line just to its left in each absorption spectrum are transitions from the ground level to each of the two $^4F_{3/2}$ Stark components. The other weak lines on the low frequency side are

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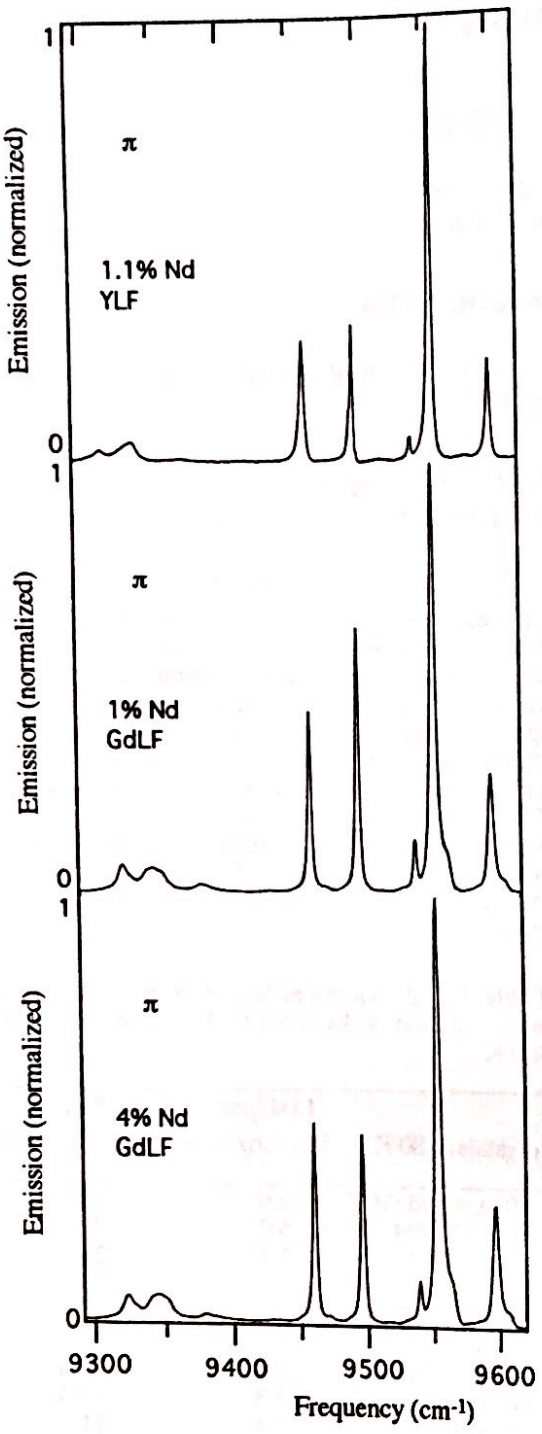

Figure 1. Relative emission spectra in π-polarization of 1.1 at.% Nd:YLF (top), 1.0 at.% Nd:GLF (middle), and 4.0 at.% Nd:GLF (bottom) at 80 K. ($^4F_{3/2} \rightarrow {}^4I_{11/2}$)

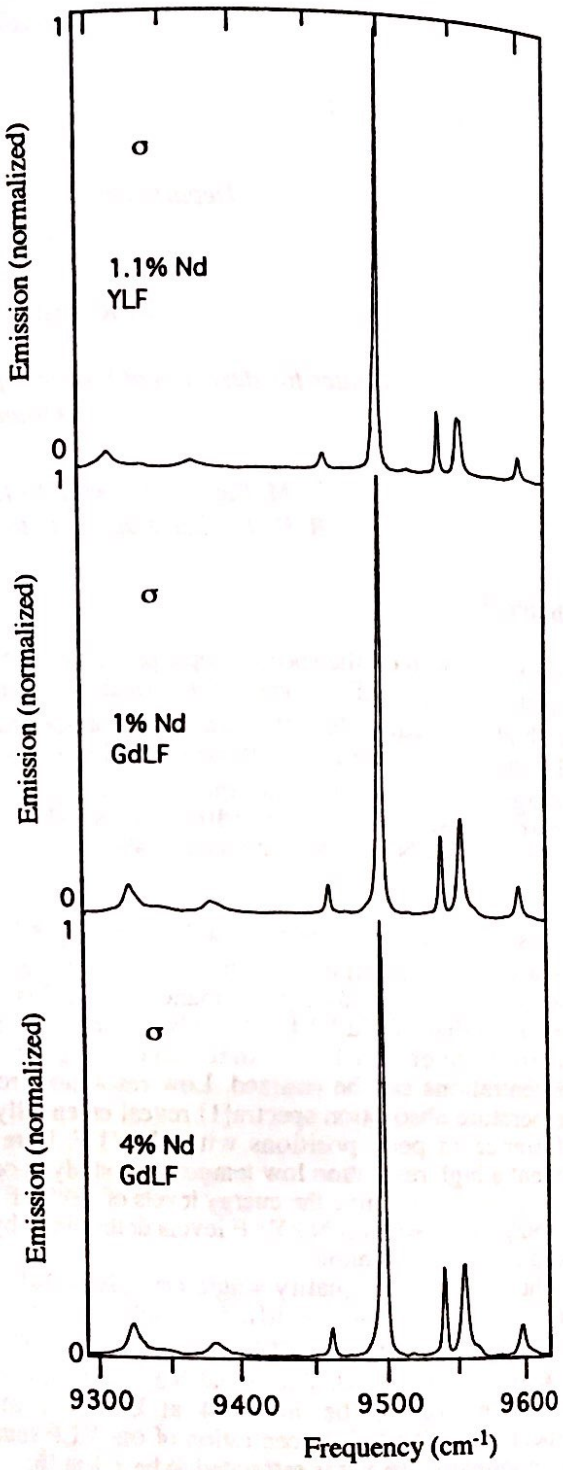

Figure 2. Relative emission spectra in σ-polarization of 1.1 at.% Nd:YLF (top), 1.0 at.% Nd:GLF (middle), and 4.0 at.% Nd:GLF (bottom) at 80 K. ($^4F_{3/2} \rightarrow {}^4I_{11/2}$)

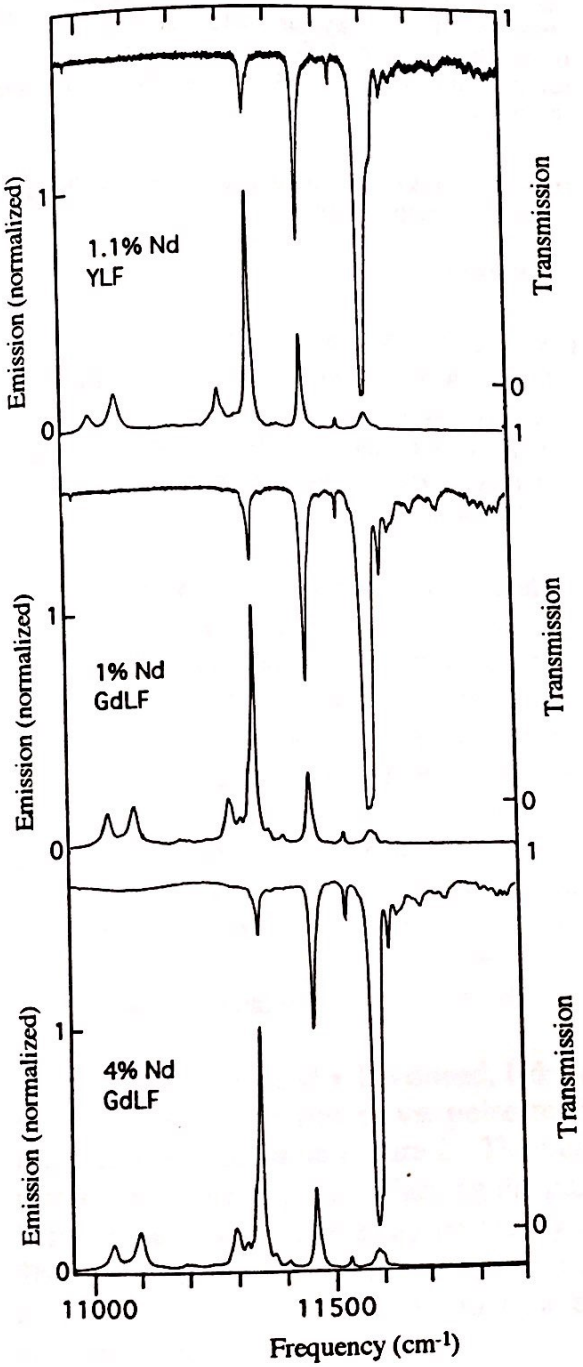

Figure 3. Transmission and relative emission spectra in π-polarization ($^4I_{9/2} \leftrightarrow {}^4F_{3/2}$) of 1.1 at.% Nd:YLF (top), 1.0 at.% Nd:GLF (middle), and 4.0 at.% Nd:GLF (bottom) at 80 K.

transitions originating in thermally populated Stark components of the ground $^4I_{9/2}$ multiplet. The weak lines on the high frequency side have comparable strengths for both Nd^{3+} concentrations and therefore are not pair lines. They can be explained as one phonon side bands. Linewidths in GLF are slightly larger than in YLF and are independent of concentration.

Fig. 4 shows the $^4I_{9/2} \rightarrow {}^4F_{5/2}, {}^2H_{9/2}$ absorption lines at a sample temperature of 80 K. The two groups of strong bands at the center of the spectra are used for diode pumping of lasers. The very close similarity in the number, spacing, relative strength, and polarization dependence of lines in each of the two materials is strong evidence that they are isostructural.

Table 2 gives those energy levels which are relevant to diode-pumped lasing. Nd:YLF has never been measured by the Fourier technique, so we present our determination of its levels. These values differ from measurements previously made by grating spectroscopy[2] by significantly more than the linewidths.

Table 2. Energy levels (cm^{-1}) of Nd^{3+} in GLF and YLF

Spectral term	GLF	YLF
$^4I_{9/2}$	0	0
	128.0	132.1
	182.0	182.5
	239.0	247.2
	496.0	526.6
$^4I_{11/2}$	1992.7	1997.1
	2036.4	2040.1
	---	2042.4
	2071.8	2077.0
	2211.4	2226.8
	2247.0	2261.9
$^4F_{3/2}$	11530.9	11535.7
	11588.7	11594.5
$^4F_{5/2}, {}^2H_{9/2}$	12534.7	12535.3
	12546.4	12544.9
	12617.9	12625.7
	12641.0	12641.8
	12656.1	12663.0
	12727.6	12729.5
	12792.7	12803.7
	12823.7	12829.4

We have measured detailed high resolution transmission and emission spectra of Nd^{3+} in a single crystal of $GdLiF_4$, a newly demonstrated laser material, for different Nd concentrations. The spectra are nearly identical to those of Nd:$YLiF_4$, strongly indicating that

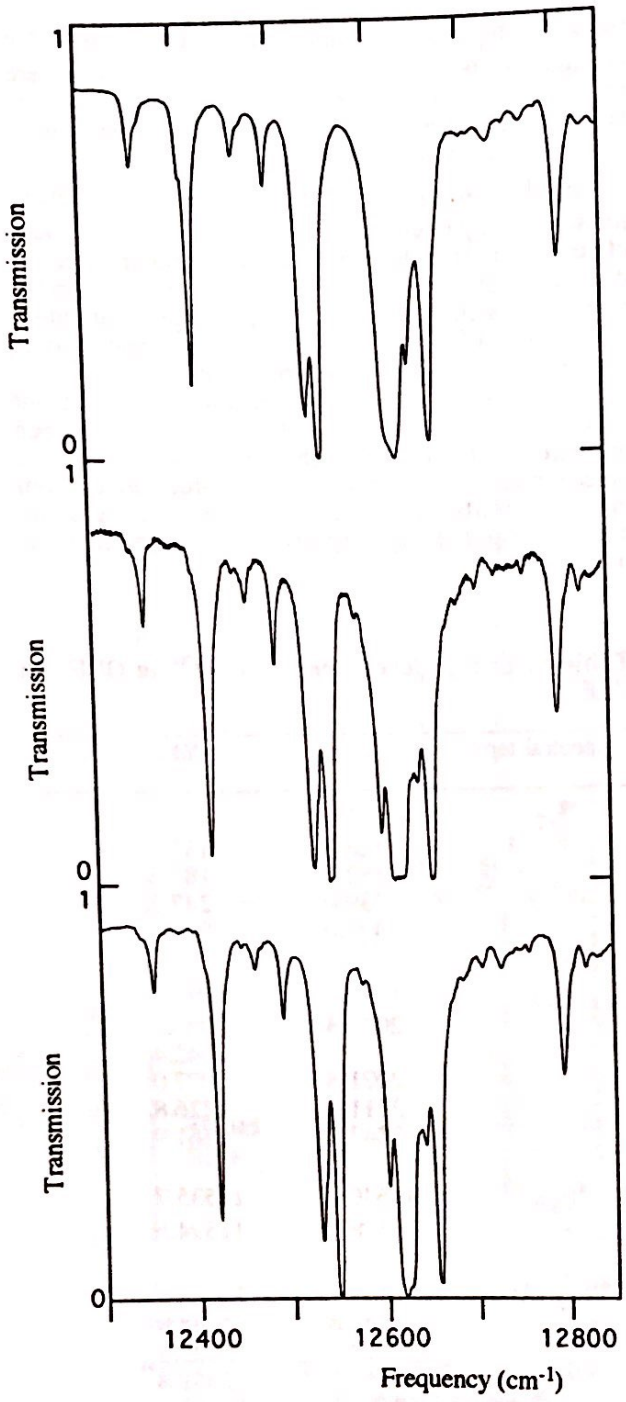

Figure 4. Transmission spectra ($^4I_{9/2} \rightarrow {}^4F_{5/2}$ and $^2H_{9/2}$) in π-polarization of 1.1 at.% Nd:YLF (top), 1.0 at.% Nd:GLF (middle), and 4.0 at.% Nd:GLF (bottom) at 80 K.

the two crystals have very similar symmetry and strength of the crystal fields and therefore are isostructural. The high dopant concentration in the GLF sample appears to effect the transitions of interest insignificantly.

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References

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